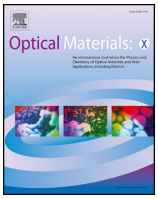
Optical Materials: X 1 (2019) 100004

Contents lists available at ScienceDirect

Optical Materials: X

journal homepage: www.journals.elsevier.com/optical-materials-x



Durability of the refractive index change induced by a single femtosecond laser pulse in glass

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ARTICLE INFO

Keywords:
Femtosecond laser writing
Refractive index change
Stability

ABSTRACT

Ultrafast-laser inscribed optical memories have been considered as a high-density low-energy-consumption alternative to the magnetization-based memories. The optical memories are based on laser-induced material modifications resulting in the refractive index change. The long-term stability of such modifications has been indicated by subjecting them to the accelerated aging by annealing at elevated temperatures. Here, the first direct evidence is provided of the durability of the type II refractive index change in BK7 glass. The investigation was performed for over 27 months at room temperature. The results show the existence of the laser pulse intensity threshold above which the magnitude of the index change does not deteriorate with time and, hence, is suitable for optical memory, photonic crystal and fibre-grating writing.

1. Introduction

Direct laser writing has emerged as an effective and flexible technique for one-step fabrication of integrated optical components in a wide range of materials [1]. Unlike smooth material processing adapted to the optical circuitry, disruptive changes in material needed for photonic crystals or optical memories rely on high index contrast or birefringence [2]. The former is achieved by point-by-point [3–6] and phase-mask [7] inscription by femtosecond laser pulses in the regime known as type II inscription. Recently, the birefringence induced by nanopatterning [8] and femtosecond pulse bursts [9] in glass has been investigated as a mechanism of ultrahigh-density optical memory. A long-term stability of femtosecond-laser induced fibre gratings and optical storage has been indicated by a series of annealing experiments performed at temperatures above 1000 K [7,8] and extrapolated to the operation temperatures down to 303 K by Arrhenius method [8]. However, a direct evidence of the durability of the femtosecond laser induced refractive index modifications at room temperature has not been reported.

Here, we address this issue and investigate spontaneous changes in the refractive index profiles of point-by-point written structures over a 27-month interval. Investigated structures are induced by tightly focused high-energy pulses suitable for memory writing. Stability has been tested against changes in the inscribed refractive index profile, namely its magnitude and dimensions. The former is of direct relevance to the memory write/read margins, while the dimensions set a limit on the memory capacity. In what follows, we describe the methods used to produce and characterise refractive index changes, report results obtained immediately after inscription and 27 months later and briefly discuss them using the established physical models of inscription.

2. Experimental method

Direct laser writing was performed by 150 fs laser pulses at 1 KHz repetition rate rendered by a Kerr mode-locked and regeneratively amplified Ti:Sapphire laser system (Spectra Physics Tsunami/Spitfire). The inscription setup is shown in Fig. 1. A part of the beam was split off into an autocorrelator for pulse duration monitoring. Pulse power was controlled by a half-wavelength plate followed by a Glan prism and measured by a thermal power meter with precision of 0.1 mW.

A BK7 glass plate sample was mounted on a 2D computer-controlled air-bearing translation stage (Aerotech A3200) extended to the third dimension (z) by a manual stage. The gaussian laser beam was tightly focused into the sample by a high-aperture 40x, NA = 0.65 microscope objective (Zeiss, Inc.). Material was processed in the form of a matrix, Fig. 1. All structures in one row were inscribed by identical single pulses, whereby the stage was moved at the speed of 20 mm/s to avoid any overlap between them, Fig. 2. The resulting distance of 20 μm between the structures was subsequently used for length calibration in x direction and 50 μm distance between the rows for calibration in y direction. Expectedly, the calibration factors matched. The laser pulse energy was changed from row to row and was varied from 0.14 μJ to

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https://doi.org/10.1016/j.omx.2019.100004
Received 15 November 2018; Received in revised form 22 December 2018; Accepted 24 December 2018
Available online 12 January 2019

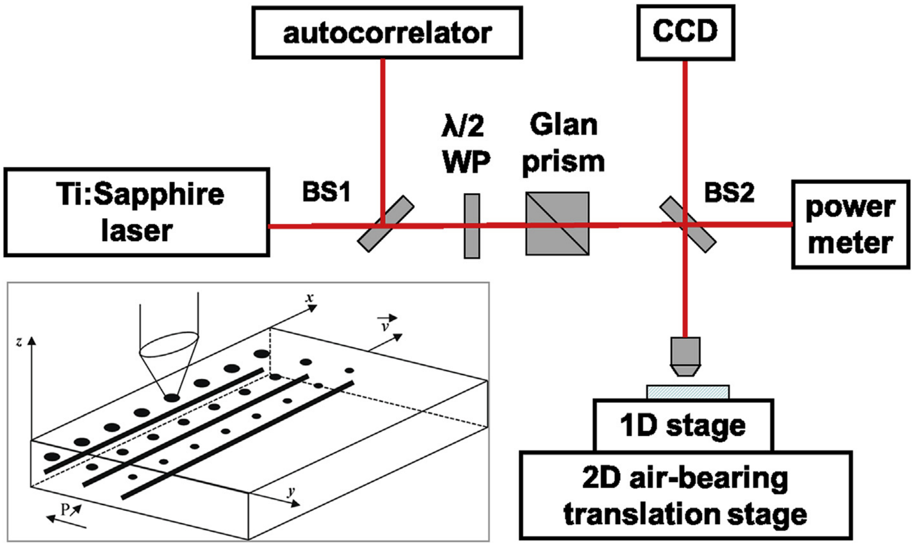

Fig. 1. Set-up for the femtosecond inscription. BS - beam splitter, WP - waveplate. Inset: Geometry of writing. Line markers correspond to rows of the structures written with the same pulse energy.

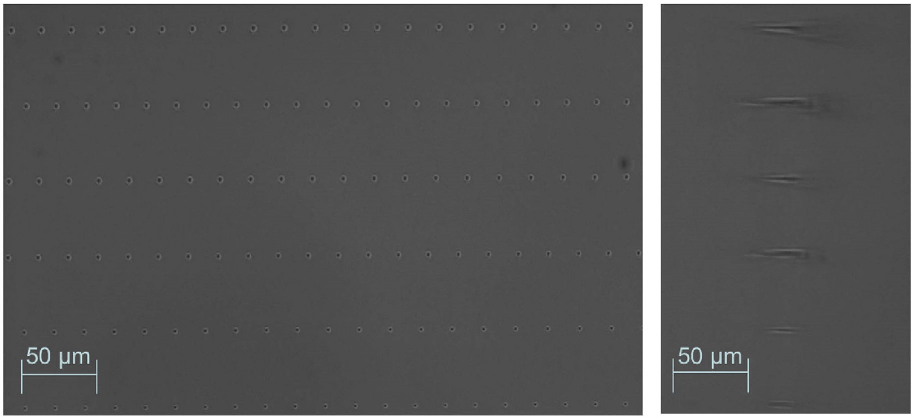

Fig. 2. a) Matrix of single-pulse traces in glass observed by a phase microscope with 20x objective in x-y plane. The rows of structures from the bottom up are inscribed with the laser pulse energies 0.41 μJ, 0.62 μJ, 1.23 μJ, 2.01 μJ, 2.63 μJ, 3.40 μJ, respectively. b) Side view of the structures, y-z plane.

3.4 μJ, Fig. 2. Each row was marked by a pre-inscribed reference line, used also for length calibration in z direction. In order to maintain the beam spot size and inscription depth during fabrication of all structures, a two-step alignment procedure was performed in three dimensions. Rough horizontal alignment was achieved by observing the reflection of an auxiliary laser beam from the mirror mounted in the sample holder during the x-y movement of the stage. Upon the sample placement, a finer alignment was achieved by repeating the procedure using the image of an unamplified Ti:Sapphire laser beam on a CCD camera. The depth of focus of 150 μm in the sample was reached by a 100μm z-stage movement in air. The reference lines were inscribed at twice greater depth.

The samples were analysed by the phase microscope Axioskop 2 MOT (Zeiss, Inc.) and the commercial software for the quantitative phase microscopy QPM (IATIA). To determine the inscribed refractive index profile, a stack of images with a structure at the focus and 1 μm displacements to both sides of it in z direction was taken by a phase microscope. The QPM software uses the transport of intensity equation to relate the recorded gradient of intensity to the change in phase [10]. The corresponding refractive index change is retrieved under the assumptions of a linear phase change and a uniform index change between any two slices, here ascertained by the small (1 μm) step in z [11]. The sample was illuminated through 20x, NA = 0.45 microscope objective (Zeiss, Inc.) Subtraction of the focal-plane image was used to correct for accidental nonuniformities in the sample illumination. The microscope light source was limited to a 20 nm bandwidth by an interference filter (Semrock). The method was previously calibrated by using optical fibres with known specifications. A detailed description of this implementation of QPM can be found in Ref. [12]. While critical to the accuracy of the method, the determination of the focal plane was not trivial due to the considerable length of the structures in the laser-propagation direction. The problem was solved by repeating the procedure along the stack and finding the region in which the index change was not dependent on the chosen focal plane. To account for pulse-to-pulse variations, refractive index change was calculated by averaging over randomly chosen structures inscribed with the same energy. In the measurement repeated after 27 months, random sampling was performed anew. The measurement error was estimated as a standard deviation of parameters of the structures within the same microscope field of view (9 per pulse energy).

Radial and axial dimensions of structures were measured with 40x, NA = 0.65 microscope objective (Zeiss, Inc.) rendering 0.168 μm/pixel. This limitation has been taken into account when analyzing and comparing results. While the index and diameter measurements did not require sample preparation, the structure length was measured upon cutting the sample in the y-z plane and polishing to the roughness of 0.1 μm. As further cutting would have disturbed the sample, statistics were not available for the structure length and its error was determined

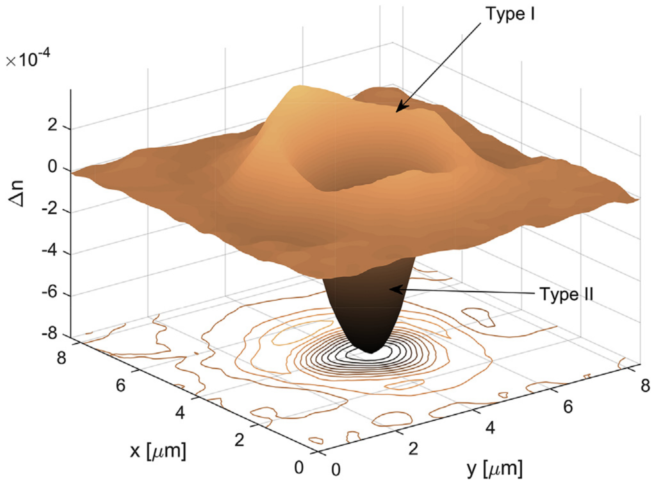

Fig. 3. Profile of a typical reconstructed refractive index change featuring modifications of both the type I and type II.

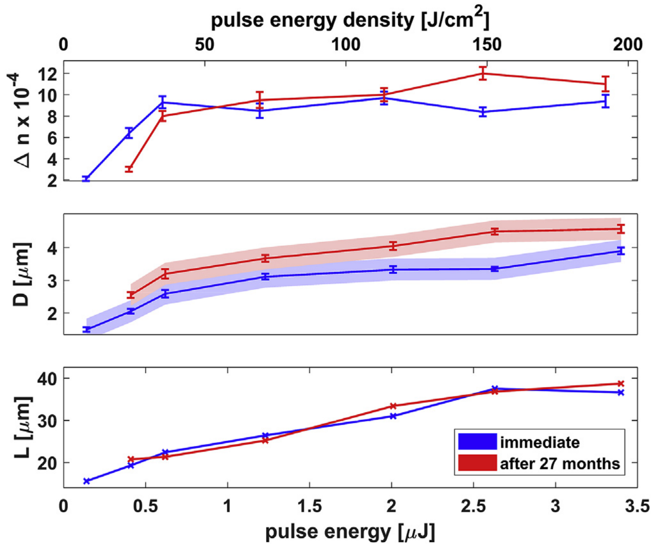

Fig. 4. Comparison of the magnitude, diameter and length of the induced refractive index change immediately after inscription (blue) and 27 months later (red). Error bars represent corresponding standard deviations for structures inscribed with the same laser pulse energy. Statistics were not available for the structure length. Shaded areas represent an error of 0.336 μm stemming from the image discretisation by the microscope camera. As this error amounts to less than 2% of the structure length, it is barely observable in the lowest graph. All lines are added to guide the eye. (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

from the pixel size. No further postprocessing was performed. The samples were stored at a room temperature for 27 months before repeating the measurement procedure.

3. Results

Inscribed structures have refractive index profile in the form of a pointed ellipsoid with the circular symmetry maintained along the beam propagation axis, Fig. 3. Estimated magnitude of the refractive index change, defined as the absolute value of the index change with respect to the substrate, was of the order of 10^{-4} and saturated at 10^{-3} for pulse energies above 1 μJ, Fig. 4. For diffraction-limited focusing, the saturation threshold pulse energy corresponds to the pulse energy density of 56.5 J/cm^2, i.e., pulse intensity of 3.8 x 10^{14} W/cm^2. Structure dimensions increased monotonously with the pulse intensity, as expected from the volume occupance by the portion of the pulse with the intensity above the inscription threshold. Standard deviations of the magnitude of the index change of 7% and the diameter of 3.5% suggest good repeatability of the inscription process.

In Fig. 4 parameters of the structures re-measured 27 months after inscription are compared with those measured immediately after inscription. Magnitude of the index change inscribed by pulses with intensities below the saturation threshold decreased over time. The decrease progressed towards lower inscription intensities to the point that the time-evolved structure inscribed with the minimum pulse intensity could not be identified by the method described. Contrarily, magnitude of the index change inscribed by pulses with above-threshold intensities withstood the test of time and increased for the highest pulse intensities. The structure diameter consistently increased irrespectively of the inscription intensity. The trend is well represented by a 23% average increase with respect to the initial diameter. The changes in structure length were negligible.

The temporal evolution of the diameter and magnitude of refractive index change are is accordance with the physical origins of the glass modification types I and II [13]. Pulse energies above the saturation point are sufficient to cause the type II change, a disruptive change in material density produced by high temperature and pressure gradients in the pulse focus. The resulting structures comprise a rarified middle region surrounded by a layer of densified material or even cracks, Fig. 3. The central change as well as cracks are robust and do not deteriorate on the time scales observed here. On the other hand, the material-stress dominated type I change at the circumference of the structure is subject to a long-term stress relaxation that may have led to the widening of the structure. It is also responsible for the observed decrease in the magnitude of the index change induced by intensities below the saturation threshold. This physical model complies with the previous investigations showing that the convergence to the final refractive index profile can be accelerated by annealing [7,14].

4. Conclusions

Durability of the refractive index change induced by single femtosecond laser pulses has been tested in a three-year long study. The measured slow evolution of the index profile confirms the stability hypothesis. The trends observed in the magnitude and dimensions of the index change induced at various laser pulse intensities correspond to the established physical models of type I and type II glass modifications. For exact quantification of the trends further studies are needed. The robustness of structures obtained in the type II regime above the saturation intensity threshold validates a long-term operation of point-by-point laser-inscribed optical memories, photonic crystals and fibre-gratings at room temperature.

Declaration of interests

None.

Acknowledgments

The author would like to thank colleagues from the Aston Institute of Photonic Technologies, Birmingham, UK for providing experimental facilities and support and to Dr. Nikola Stojanovic for useful discussions. The work has been supported by the Ministry of Education, Science and Technological Development of Serbia under the grant No. III45010 and European Union's Horizon 2020 research and innovation programme under the Marie Sklodowska-Curie grant agreement No. 691051.

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